

INVESTIGATIONS OF THE ROLE OF COAL THERMOPLASTIC PROPERTIES AND COKE STRUCTURE IN GASIFICATION PROCESSES

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INTRODUCTION

Coal classification schemes have been developed over many years from studies of the physical and chemical properties of coals as a method of dividing coals into various categories according to their behaviour during carbonisation and combustion. Although there is no universally accepted scheme, most coal classification schemes are based on two main types of parameters:- a) volatile matter, and b) caking and swelling properties. The limited scope and empirical nature of the techniques used in classification schemes and their variation with experimental conditions means that classification schemes have only limited applicability when comparing coal behaviour in high pressure gasifiers where the nature of the process is a primary consideration and the experimental conditions are vastly different from those used in the tests.

The thermoplastic properties of coal, as well as characterising the initial agglomeration processes, are important in determining both porous structure and carbon texture of the coke product. Both the coal and coke structures are significant factors in determining gasification behaviour. The coke strength and reactivity during gasification are of considerable significance in fixed bed gasifiers. The agglomerated coke is broken up by the stirrer to facilitate the countercurrent flow of gases and solids(1) and hence, the mechanical strength of the coke may be an important factor. In the case of weak cokes, a wide size range may be produced and increased levels of dust may occur causing a loss of efficiency and throughput. Coke structure is important in determining reactivity towards oxidising gases which is a main gasification step.

The objective of this study is to provide information on the dependence of coal and coke properties under experimental conditions found in fixed bed gasifiers. The direct link between measurements of coal thermoplasticity and coke structure obtained by examining the structure of the carbonised residues from the high pressure thermoplasticity measurements is considered to be an important aspect of the investigation since it eliminates any possible ambiguities in comparing various sets of data. From these considerations and other experiments under simulated gasifier conditions, comparisons with gasifier operational data can be made which should make it possible to develop optimum methods for predicting coal performance in a particular process.

EXPERIMENTAL

The coals used in this study were a strongly caking coal (Coal Oa, NCB Classification Scheme 301a) and a moderately caking coal (Coal H1, NCB Classification 602) with the characteristics given in table 1.

The high pressure dilatometer used in this study has been described, in detail, elsewhere(2). It operates at fixed pressures up to 10MPa and heating rates up to $60^{\circ}\text{Cmin}^{-1}$. In all other respects, the dilatometer geometry and sample preparation were standard (BS1016:Part12) with the exception of the preliminary temperature stabilisation which was not used with heating rates above $3^{\circ}\text{Cmin}^{-1}$. The standard deviations of the contraction and dilatation values are estimated to be 5-10% and 10-15% respectively.

The high pressure plastometer used in this investigation has a geometry which is a scaled down version (x50%) of the Brabender Plastometer which has been described previously(3). It operates at fixed pressure up to 10MPa, heating rates up to $40^{\circ}\text{Cmin}^{-1}$ and rotational speeds in the range 2-30 revs per minute. A rotational speed of 10 revs per minute and a sample weight of 12.5g (particle size fraction 0.5-1.0mm) were used in all experiments.

The carbonised residues from the high pressure dilatometer and plastometer (HTT 550C, Soak Time 0.1hour) were crushed, mixed with epoxy resin and mounted in the form of discs. These discs were ground and polished. The polished blocks were examined by polarised light microscopy with a X50 objective at an overall magnification of X500 with the polars adjusted close to extinction. The different structural features were classified according to their appearance, size and shape, in terms of mosaic anisotropy of various grain size, flow type anisotropy and isotropic material. A 300 point count was used for quantitative measurements on each sample. The error in these measurements is estimated to be better than $\pm 5\%$. The Optical Anisotropy Index (OAI) was calculated according to the equation described previously (4,5) which is given below.

$$\text{OAI} = 1 \cdot \text{I} + 2 \cdot \text{f} + 3 \cdot \text{m} + 4 \cdot \text{c} + 5 \cdot \text{gf} + 6 \cdot \text{f} + 7 \cdot \text{b}$$

where f=fine mosaic, m=medium mosaic, c=coarse mosaic, gf=granular flow, f=flow and b=basic anisotropy.

RESULTS AND DISCUSSION

Figure 1 shows a comparison of the plastometry results obtained for Coal H1 with a heating rate of $3^{\circ}\text{Cmin}^{-1}$ at pressures of 0.5 and 5MPa. It is clear that a change in pressure has modified the plastometry characteristics. For this particular coal increasing pressure causes a decrease in the caking intensity while the caking intensity remains virtually unchanged. It is interesting to note that the onset of caking is sharper at lower pressure with the initial softening temperature decreasing markedly with increase in pressure. The plastic range also increases correspondingly with increase in pressure. A graph of plastometry parameters against pressure is shown in Figure 2. This clearly illustrates the different behaviour of the caking

and coking intensities with respect to pressure. In contrast, the plastometry parameters of coal Oa show similar trends with the exception of the coking intensity which does not change markedly with changes in pressure. The effect of heating rate on the plastometry characteristics of both coals is quite marked. The plastometry curves show that the caking intensity for both coals is not affected greatly by changes in the heating rate and this is similar to the effect of changes in carbonisation pressure. Figure 3 shows that both the coking and maximum fluidity intensities decrease dramatically with increasing heating rate. The apparent plastic range also increases with heating rate.

To understand the changes in plastometry characteristics, the swelling during carbonisation needs to be considered. Previous work has shown(2,4-8) that the swelling characteristics of coals are markedly dependent on heating rate and pressure, and cannot be predicted accurately from their swelling under standard conditions(atmospheric pressure and low heating rate). The three types of dilatation behaviour with respect to pressure which have been observed, depend to some extent on coal rank and are as follows:

- a) an increase in dilatation with pressure;
- b) an increase to a maximum at 1-1.5MPa followed by a decrease;and
- c) a decrease in dilatation with pressure.

In contrast, the dilatation of coals as measured by the method used in this study, increases with increasing heating rate. A limiting value of the dilatation was observed at a heating rate of $20^{\circ}\text{Cmin}^{-1}$ in a previous study(4). An increase in pressure will have two main effects:a) a reduction in the volume of gas trapped in the fluid phase which would tend to decrease the swelling; and b) an increase in the secondary reactions by decreasing the volatility of tar and increasing the residence time in the fluid phase which would tend to increase dilatation. The changes in dilatation behaviour can be rationalised by considering the balance between the release of gaseous and liquid volatiles. The variation of the dilatation of coal Oa with heating rate at 2MPa pressure is shown in Figure 4. The dilatation of coal Oa increases with heating rate but decreases with increase in pressure(0.5-5MPa). A graph of dilatation against plastometry torque parameters for a range of heating rates($3-20^{\circ}\text{Cmin}^{-1}$) with a carbonisation pressure of 2MPa is shown in Figure 5. This graph clearly shows that there is a correlation between some of the plastometry and dilatometry parameters. It is noticeable that the caking intensity is not as sensitive to changes in dilatation as the coking and maximum fluidity intensities which change significantly.

Investigations of the optical anisotropic content of cokes derived from a wide range of coals have shown that it is a useful parameter for characterising their structure. Previous work has shown(4,5,9) that changes in carbonisation pressure and heating rate can have a considerable effect on the optical anisotropic

content of cokes. In particular, the anisotropic content of cokes usually increases substantially with pressure at heating rates of $40^{\circ}\text{Cmin}^{-1}$ to a limiting value above approximately 3MPa. The corresponding increase in anisotropic content appears to be much smaller at low heating rates. However, there is only a limited amount of information available in the literature on this aspect. In general, the change in OAI development in relation to pressure varies with rank in a fairly systematic manner but there is no direct correlation between individual rank and anisotropy parameters. The results suggested that the OAI changes are related to the influence of pressure on the retention of plastic material and modification of the plastic range. The variation of optical anisotropy index(OAI) for cokes derived from coal Oa at a carbonisation pressure of 2MPa with heating rate is shown in Figure 6. It is apparent that the OAI increases with increasing heating rate. For this particular coal, the changes in OAI also correlate with some of the plastometry parameters, in particular, the coking intensity, the intensity at the point of maximum fluidity and the plastic range. Further work is needed on a wide range of coals to establish definitive correlations between plastometry parameters, swelling characteristics and coke porosity and structure.

CONCLUSIONS

The relationship between coal thermoplastic properties and coke structure is very complex. The results of this investigation have shown that these properties are strongly dependent on the experimental conditions and, in particular:

- a) increased swelling is accompanied by a decreased plastometry coking intensity.
- b) The plastometry caking intensity is considerably less sensitive to change in experimental conditions than the other plastometry parameters.
- c) The optical anisotropy index is increased by heating rate and this is accompanied by changes in plastic range, plastometry coking intensity and a decrease in the apparent viscosity at the point of maximum fluidity.

These changes can be rationalised by considering the release of gaseous and liquid volatiles during the carbonisation of the coal. The lack of sensitivity of the caking intensity to heating rate and pressure is consistent with its origin being partly softening prior to the major decomposition. Further work is needed to relate coal thermoplasticity measurements to other coke structural data, in particular, texture and porosity measurements.

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Table 1
Characterisation Data for the Coals used in this Study

	Coal Oa	Coal H1
Rank	301a	602
Proximate Analysis(wt%dry basis)		
Volatile	18.9	38.7
Matter		
Ash	9.2	3.8
Ultimate Analysis(wt%dry basis)		
Carbon	82.6	79.6
Hydrogen	4.3	5.3
Chlorine	0.05	-
Sulphur	0.86	1.65
Caking and Swelling Properties		
Swelling Number	6	3
Dilatometry		
Dilatation(%)	41	36
Contraction(%)	23	30
Plastic Range(°C)	87	73
Gieseler Plastometry		
Max.Fluidity(ddpm)	590	535
T ₃ -T ₁ (°C)	91	69
Petrographic Analysis		
Reflectance(%)	1.46	0.71
Maceral Analysis(vol.%)		
Vitrinite	65	78
Exinite	-	9
Inertinite	35	13

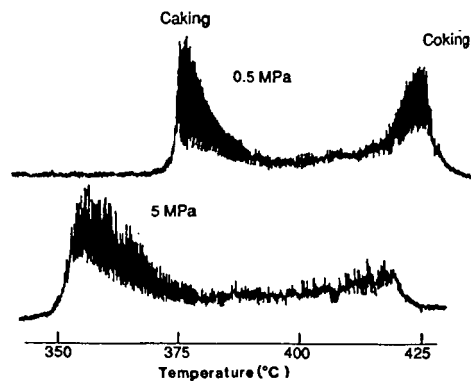


Fig. 1. A comparison of the plastometry curves for coal H1 at 0.5 and 5 MPa (heating rate $3^{\circ}\text{C min}^{-1}$)

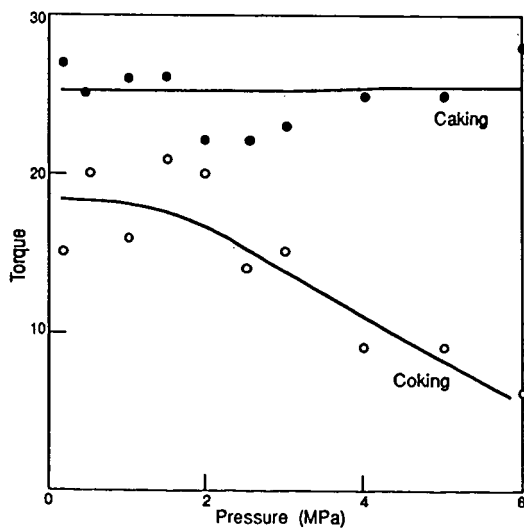


Fig.2. The variation of plastometry torque parameters with pressure for Coal H1 (Heating Rate $3^{\circ}\text{C min}^{-1}$)

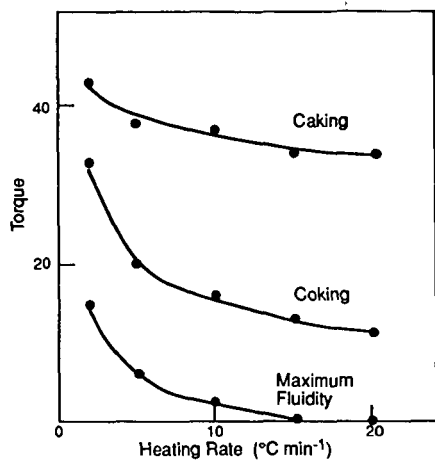


Fig.3. The variation of plastometry torque parameters with heating rate for coal H1 (Pressure 2 MPa)

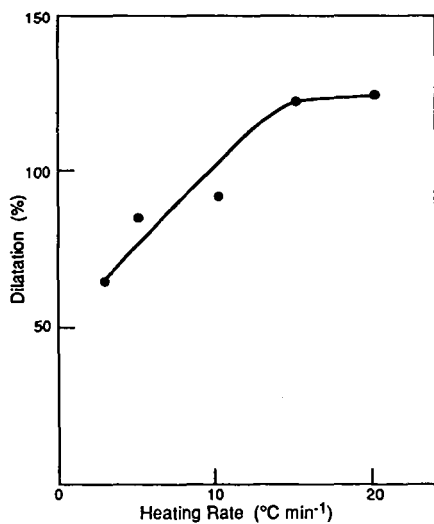


Fig.4. The effect of heating rate on the dilatation of coal Oa (Gauge pressure - 2 MPa)

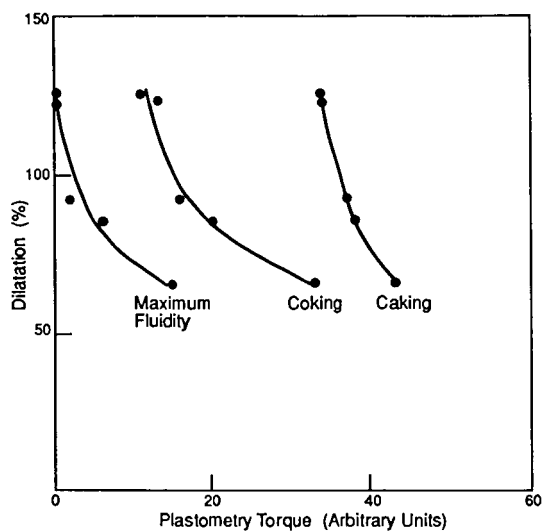


Fig.5. The variation of dilatation with plastometry torque parameters at 2 MPa pressure for coal Oa

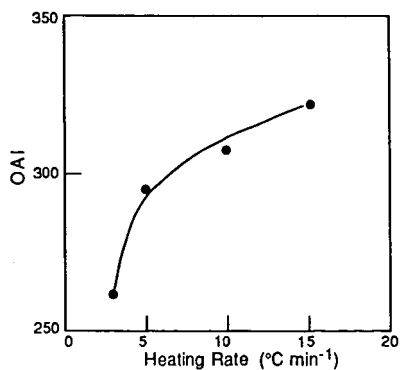


Fig.6. The variation of optical anisotropy index (OAI) with heating rate for coal Oa (Pressure 2 MPa)